

# Coherent energy scale revealed by ultrafast dynamics of $UX_3$ ( $X=\text{Al, Sn, Ga}$ ) single crystals

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Temperature dependence of relaxation dynamics of  $UX_3$  ( $X = \text{Al, Ga, Sn}$ ) compounds is studied using time resolved pump-probe technique in the reflectance geometry.  $UGa_3$  is an itinerant anti-ferromagnet, while  $UAl_3$  and  $USn_3$  are spin fluctuation systems. For  $UGa_3$ , our data are consistent with the formation of a spin density wave SDW gap as evidenced from the quasidivergence of the relaxation time  $\tau$  near the Néel temperature  $T_N$ . For  $UAl_3$  and  $USn_3$ , the relaxation dynamics shows a change from single exponential to two exponential behavior below a particular temperature, suggestive of coherence formation of the  $5f$  electrons with the conduction band electrons. This particular temperature can be attributed to the spin fluctuation temperature  $T_{sf}$ , a measure of the strength of Kondo coherence. Our  $T_{sf}$  is consistent with other data such as resistivity and susceptibility measurements. The temperature dependence of the relaxation amplitude and time of  $UAl_3$  and  $USn_3$  were also fitted by the Rothwarf-Taylor model. Our results show ultrafast optical spectroscopy is sensitive to  $c$ - $f$  Kondo hybridization in the  $f$ -electron systems.

## I. INTRODUCTION

The uranium compounds  $UX_3$ , where  $X$  is a IIIb (Al, Ga, In, Tl) or IVb (Si, Ge, Sn, Pb) element, crystallize in the cubic  $AuCu_3$ -type structure<sup>1</sup> and have U-U distances ( $d_{U-U}$ ) much larger than the Hill limit ( $\sim 3.5$  Å) for uranium compounds.<sup>2</sup> The different degree of hybridization of the  $5f$  electron orbitals with the conduction electron orbitals in these compounds leads to a wide range of magnetic behavior such as Pauli enhanced paramagnetism ( $UAl_3$ ,  $USi_3$ , and  $UGe_3$ ), antiferromagnetism ( $UGa_3$ ,  $UPb_3$  and  $UIn_3$ ), and heavy fermion behavior ( $USn_3$ ).<sup>1,3,4</sup> Due to the the above-mentioned properties and the availability of high quality crystals,  $UX_3$  compounds are ideal candidates for studying how physical properties and underlying electronic structure are related.

The anomalous behavior of the resistivities of  $UX_3$  compounds can be explained on the basis of spin fluctuations in narrow  $5f$  bands.<sup>5,6</sup> A temperature characteristic of the spin fluctuations in the  $UX_3$  compounds is the spin fluctuation temperature,  $T_{sf}$ , which expresses the strength of hybridization between  $f$  and conduction electrons ( $c$ - $f$  hybridization). The degree of hybridization is related to the degree of delocalization of the  $f$ -electrons. A high value of  $T_{sf}$  corresponds to more easily hybridized (delocalized) electrons. Above  $T_{sf}$ ,  $f$ -electrons are localized; whereas below  $T_{sf}$ , there is quasiparticle coherence from the hybridization between  $f$ -electrons and conduction electrons, *i.e.*,  $f$ -electrons now become more delocalized (or itinerant). The effective hybridization below  $T_{sf}$  leads to changes in measured physical properties. For example, the electrical resistivity changes from a  $T$ -linear law above  $T_{sf}$  to a  $T$ -quadratic law below this temperature.<sup>6-9</sup> The temperature at which the magnetic susceptibility reaches a Curie-Weiss law is theoretically

of the order of  $T_{sf}$ .<sup>6</sup> A modified Curie-Weiss law, *i.e.*  $\chi(T) = \chi_0 + C/(T + T^*)$ , associates  $T^*$  with  $T_{sf}$  for relatively strong  $c$ - $f$  hybridization.<sup>10,11</sup>

Ultrafast time-resolved pump-probe spectroscopy has been recognized as a powerful technique to study the nonequilibrium carrier dynamics in strongly correlated electron materials. In addition to distinguishing different phases in a material by their different relaxation dynamics, it can discern whether one phase coexists or competes with another phase in close proximity,<sup>12,13</sup> giving information on the nature of low energy electronic structure of correlated electron systems, for example, in high-temperature superconductors. Pump-probe experiments have also been performed on actinide compounds, such as the itinerant antiferromagnets  $UNiGa_5$  and  $UPtGa_5$ ,<sup>14,15</sup> and the heavy-fermion superconductor  $PuCoGa_5$ .<sup>16</sup>

The hybridization between the conduction electrons and the localized  $f$  electrons also causes a narrow gap to form in the density of states near the Fermi level.<sup>17</sup> This gap, called the hybridization gap, results in a relaxation bottleneck, evidenced by an increase in the relaxation time  $\tau$  at low temperatures. For example, in heavy fermions such as  $YbAgCu_4$  and  $SmB_6$ ,  $\tau$  increases monotonically with decreasing temperature.<sup>17</sup> The temperature dependence of the relaxation amplitude and time were fit using the Rothwarf-Taylor (RT) model. In this paper, we investigate the ultrafast dynamics in three isostructural uranium compounds,  $UAl_3$ ,  $UGa_3$  and  $USn_3$ , using the ultrafast pump-probe technique. The variation in hybridization strength is responsible for the differences in properties of these three isostructural compounds.  $UAl_3$  and  $USn_3$  are categorized as spin-fluctuation systems.<sup>7,18-22</sup>  $UGa_3$  does not behave as a spin fluctuation system, but is an itinerant  $5f$  electron antiferromagnet. In fitting the transient change in reflectivity for  $UAl_3$  and  $USn_3$ , we needed a two-exponential decay function below  $T_{sf}$ , which points to the presence of

two relaxation channels below  $T_{sf}$ . This arises from the hybridization between  $f$  electrons and conduction electrons below  $T_{sf}$ . This shows that the ultrafast pump probe technique is sensitive to  $c$ - $f$  hybridization in  $f$ -electron systems. Our  $T_{sf}$  is consistent with that obtained from resistivity and susceptibility measurements. We were also able to fit the temperature dependence of the relaxation amplitude and time using the RT model. For  $\text{UGa}_3$ , the relaxation time diverges as the temperature approaches the Néel temperature  $T_N$ , corresponding to the formation of a spin density wave (SDW) gap near the Fermi level.

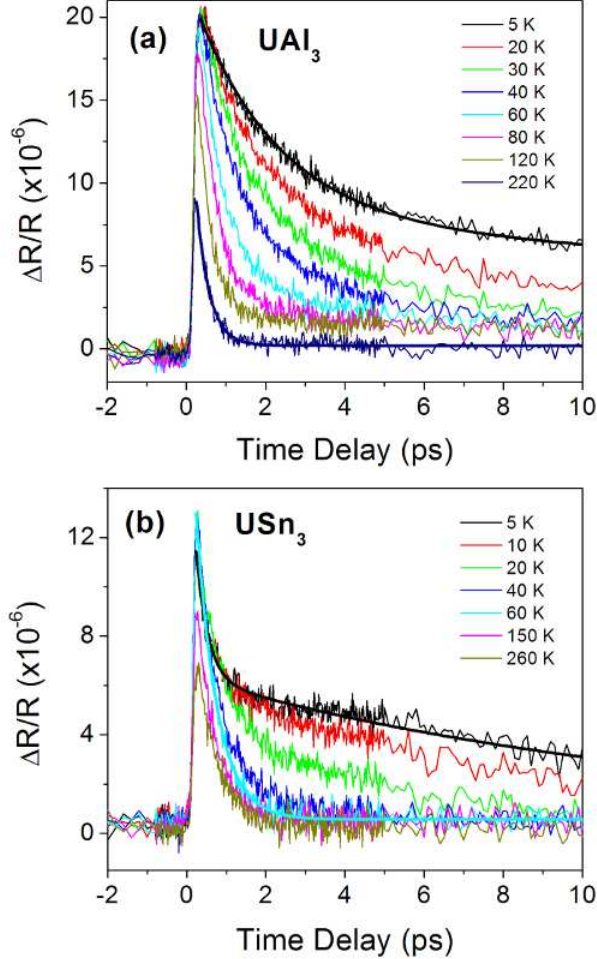


FIG. 1: Transient reflection  $\Delta R/R$  versus pump-probe time delay at different temperatures for (a)  $\text{UAl}_3$ , and (b)  $\text{USn}_3$ . Thick solid curves denote exponential fits of data.

## II. EXPERIMENTAL SETUP

In our pump-probe experimental setup in reflectance geometry, a Ti:sapphire laser producing sub-100 fs pulses at  $\approx 800$  nm (1.55 eV) was used as a source of both pump and probe pulses. The pump and probe pulses were cross

polarized. The pump spot diameter was  $60 \mu\text{m}$  and that of probe was  $30 \mu\text{m}$ . The reflected probe beam was focused onto an avalanche photodiode detector. The photoinduced change in reflectivity ( $\Delta R/R$ ) was measured using lock-in detection. In order to minimize noise, the pump beam was modulated at 100 kHz with an acousto-optical modulator. The experiments were performed with an average pump power of 2 mW, giving a pump fluence of  $\sim 1 \mu\text{J}/\text{cm}^2$ . The probe intensity was approximately ten times lower. Data were taken from 10 K to 300 K. The experiments were performed on single crystals of  $\text{UX}_3$  ( $X = \text{Al}, \text{Ga}, \text{Sn}$ ) grown using standard flux technique, with X used as the flux in each case.<sup>22</sup>

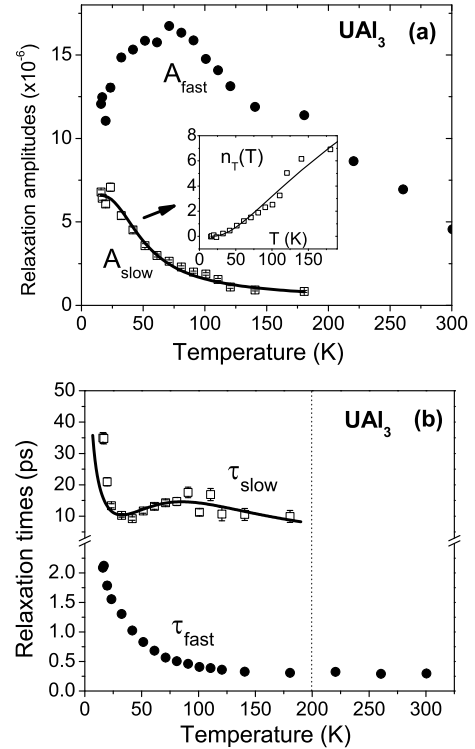


FIG. 2: Temperature dependence of (a) amplitudes and (b) relaxation times for  $\text{UAl}_3$ . Solid lines are fits to the RT model of the slow component.

## III. $\text{UAl}_3$ AND $\text{USn}_3$

In Fig. 1 we show the  $\Delta R/R$  at different temperatures for (a)  $\text{UAl}_3$  and (b)  $\text{USn}_3$ , as a function of the time delay between the pump and probe pulses. In both  $\text{UAl}_3$  and  $\text{USn}_3$ , only a fast relaxation of  $\sim 500$  fs, which is typical of regular metals, is observed at high temperatures. At low temperatures, an additional slow, positive picosecond relaxation is observed. Data at low temperatures are fit-

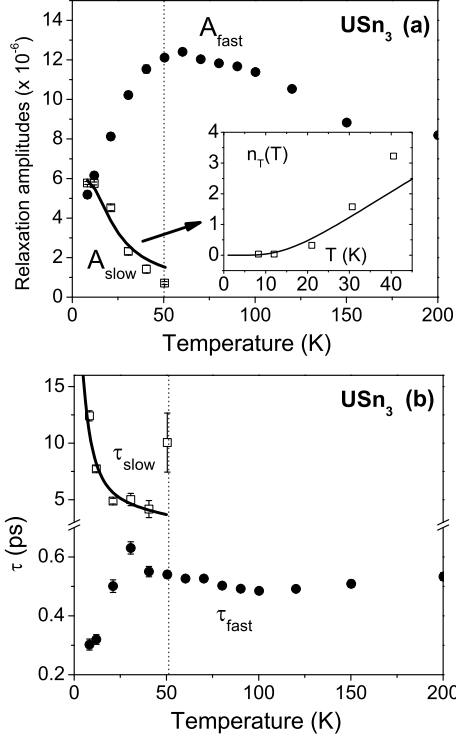


FIG. 3: Temperature dependence of (a) amplitude and (b) relaxation time for  $\text{USn}_3$ . Solid lines are fits to the RT model of the slow component.

ted to the two-exponential decay function  $\Delta R/R(t) = A_{fast}(T) \exp(-t/\tau_{fast}) + A_{slow}(T) \exp(-t/\tau_{slow})$ . This change from one- to two-exponential decay occurs at a particular crossover temperature —  $\sim 200$  K for  $\text{UAl}_3$  and  $\sim 50$  K for  $\text{USn}_3$ , suggestive of two relaxation channels below this crossover temperature. These crossover temperatures are of the order of the spin fluctuation temperatures  $T_{sf}$  obtained in these compounds from temperature-dependent electrical resistivity and magnetic susceptibility measurements ( $\sim 150$  K for  $\text{UAl}_3$ <sup>6,23</sup> and  $\sim 50$  K for  $\text{USn}_3$ <sup>6,24,25</sup>) We thus associate this crossover temperature to the spin fluctuation temperature  $T_{sf}$ .

To understand the different characteristic temperatures in  $\text{UAl}_3$  and  $\text{USn}_3$ , we have also performed band structure calculations in the framework of the density functional theory, by using the WIEN2k linearized augmented plane wave method.<sup>26</sup> A generalized gradient approximation<sup>27</sup> was used to treat exchange and correlation. Spin-orbit coupling was included in a second-variational way. The obtained U partial 5f density of states, as shown in Fig. 4, indicates a narrower peak width near the Fermi energy, in  $\text{USn}_3$  as compared with  $\text{UAl}_3$ . In addition, one can see that the splitting between the two major peaks is smaller in  $\text{USn}_3$  than in  $\text{UAl}_3$ . In

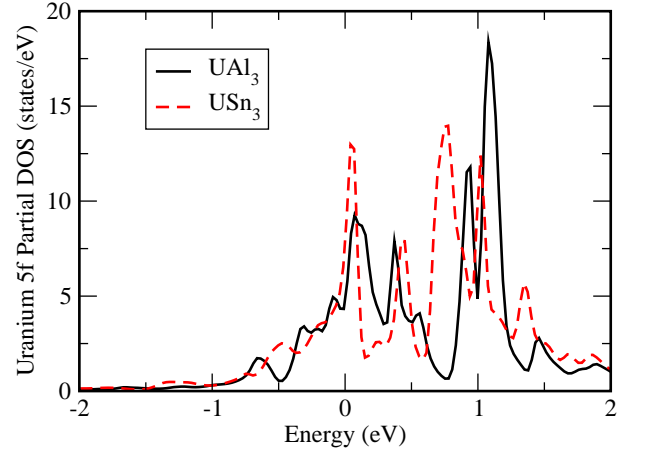


FIG. 4: Total DOS calculated from the LAPW method for  $\text{UAl}_3$  and  $\text{USn}_3$ , in the magnetic unit cell, in the energy range  $(-2, 2)$  eV. Note the narrower peak width near the Fermi energy ( $E=0$ ) in  $\text{USn}_3$  compared to  $\text{UAl}_3$ .

view of the fact that the spin-orbit coupling is quite local to the U atoms, one would expect the same effect on both  $\text{USn}_3$  and  $\text{UAl}_3$ . A reasonable explanation for this difference is a smaller hybridization gap in  $\text{USn}_3$  compared to  $\text{UAl}_3$ , due to the weakening of the hybridization in  $\text{USn}_3$  — a result of the lattice expansion ( $a=4.626$  Å in  $\text{USn}_3$  versus  $a=4.264$  Å in  $\text{UAl}_3$ ).<sup>23</sup> Though conventional band structure calculations underestimate the correlation effect, the trend of smaller coherence energy scale in  $\text{USn}_3$  than in  $\text{UAl}_3$  should be robust, as has recently been exemplified in other isostructural actinide compounds.<sup>28</sup>

In this context, the two-exponential behavior at low temperature can be explained by the  $c$ - $f$  hybridization occurring below  $T_{sf}$ . Below  $T_{sf}$ , the interaction of partially-filled  $f$  shell electrons with conduction electrons lead to the formation of heavy quasiparticles.<sup>29</sup> As the  $f$ -electrons are localized above  $T_{sf}$ , relaxation occurs through phonon channel only. Hence only a single exponential decay is expected above  $T_{sf}$ . When  $T < T_{sf}$ , the spin fluctuation channel opens up due to hybridization. Electrons now relax via *both* phonon and spin fluctuation channels resulting in a two-exponential decay behavior. Also, a higher  $T_{sf}$  value in  $\text{UAl}_3$  compared to  $\text{USn}_3$  points to a stronger  $c$ - $f$  hybridization, which is expected, as  $c$ - $f$  hybridization tends to decrease as the size of the non- $f$  atom increases,<sup>3,30</sup> which causes the lattice expansion as we discussed above.

The hybridization between the conduction band and the localized  $f$ -levels also results in the formation of a narrow gap in the density of states near the Fermi level, called the hybridization gap. The presence of this gap causes a bottleneck in quasiparticle relaxation, resulting in a divergence of the relaxation time at low temperatures. The temperature dependence of the relaxation amplitude and relaxation time can be quantitatively explained by the Rothwarf-Taylor (RT) model. It is a phenomenological model that was used to describe the re-

laxation of photoexcited superconductors,<sup>31,32</sup> itinerant antiferromagnets<sup>14,15</sup> and heavy-fermion metals,<sup>17</sup> where the presence of a gap in the electronic density of states gives rise to a relaxation bottleneck for carrier relaxation. In heavy fermions, after the initial photo-excitation by a pump pulse, the subsequent fast relaxation due to electron-electron scattering results in excess densities of electron-hole pairs (EHPs) and high-frequency phonons (HFPs). When an EHP with energy  $\geq \Delta$  ( $\Delta$ = hybridization gap) recombines, a HFP is created. The HFPs released in the EHP recombination are trapped within the excited volume and can re-excite EHPs; hence they act as a bottleneck for EHP recombination, and recovery is governed by the decay of the HFP population. The evolution of EHP and HFP populations is described by a set of two coupled nonlinear differential equations.

The results of the RT model are as follows:<sup>17,33</sup> from the amplitude  $A(T)$ , one obtains the density of thermally excited EHPs  $n_T$  via the relation

$$n_T(T) \propto \mathcal{A}(T)^{-1} - 1 \quad (1)$$

where  $\mathcal{A}(T)$  is the normalized amplitude ( $\mathcal{A}(T) = A(T)/A(T \rightarrow 0)$ ). Then we fit the experimental  $n_T(T)$  to the expression<sup>17</sup>

$$n_T(T) \propto \sqrt{T} \exp(-\Delta/T), \quad (2)$$

where the hybridization gap  $\Delta$  is temperature independent (or very weakly temperature dependent) and can be obtained from the fitting. Moreover, for a constant pump intensity, the temperature-dependence of  $n_T$  also governs the temperature-dependence of  $\tau^{-1}$ , given by

$$\tau^{-1}(T) = \Gamma[\delta(\beta n_T + 1)^{-1} + 2n_T](\Delta + \alpha T \Delta^4) \quad (3)$$

where  $\Gamma$ ,  $\delta$ ,  $\beta$  and  $\alpha$  are  $T$ -independent fitting parameters.

Since, below  $T_{sf}$ , the second relaxation component appears, we attribute it to relaxation across the hybridization gap, and use the RT model to fit the its amplitude and relaxation time below  $T_{sf}$  in  $\text{UAl}_3$  and  $\text{USn}_3$ . The inset of Fig. 2(a) shows  $n_T(T)$  obtained from  $A_{slow}(T)$  using Eq. (1), with the solid line being the fit to Eq. (2), with the fitting parameter  $\Delta \approx (230 \pm 10)$  K. The fitted values of  $n_T(T)$  are then inserted into Eq. (3) to fit the experimental values of  $\tau_{slow}(T)$ , shown in Fig. 2(b). Similar fits are also done for  $\text{USn}_3$ , as shown in Fig. 3, yielding  $\Delta \approx (90 \pm 20)$  K. The good fits show that the slow relaxation component in both  $\text{UAl}_3$  and  $\text{USn}_3$  can be described by assuming EHPs relaxing across the hybridization gap near the Fermi surface. More interestingly, the extracted hybridization gap in  $\text{UAl}_3$  is larger than in  $\text{USn}_3$ , in qualitative agreement with the band structure results. This comparison of hybridization gap is also consistent with that of spin fluctuation energy scale  $T_{sf}$  discussed above. Our results show that the ultrafast pump-probe technique is sensitive to the hybridization of  $f$ -electron orbitals with the conduction electron orbitals.

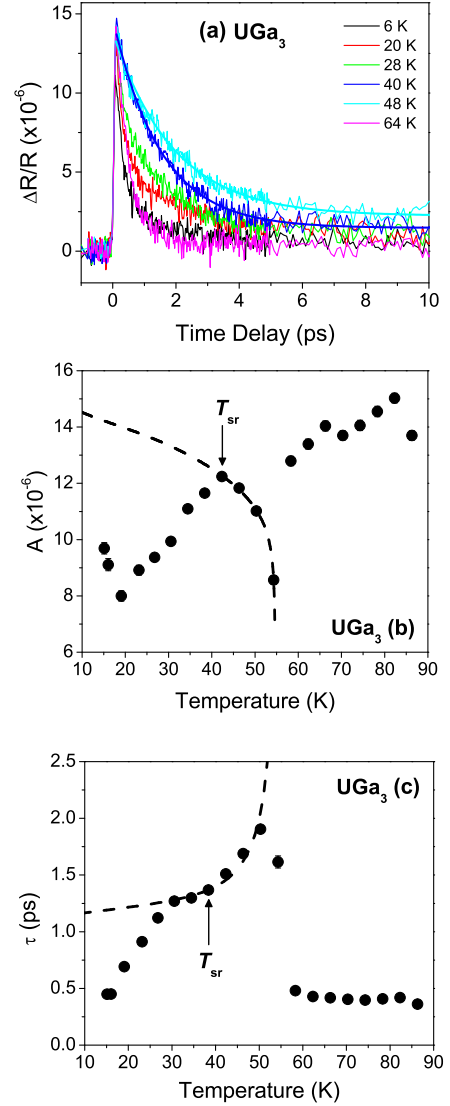


FIG. 5: (a) Photoinduced change in reflectivity  $\Delta R/R$  versus pump-probe time delay at different temperatures of  $\text{UGa}_3$ . Thick blue (cyan) curves denote one-exponential fits of data at  $T=40$  K (48 K). Temperature dependence of (b) amplitude and (c) relaxation time for  $\text{UGa}_3$  obtained from one-exponential fits, with dashed lines in (b) and (c) being fits to the RT model from 40 K to  $T_N$ .

#### IV. $\text{UGa}_3$

We now turn to the relaxation dynamics of  $\text{UGa}_3$ .  $\text{UGa}_3$  is not a spin fluctuation system — it is a SDW system with Néel temperature  $T_N=65$  K. It is a moderate heavy fermion with Sommerfeld coefficient 52 mJ/K<sup>2</sup>.mol,<sup>34</sup> and is reported to follow a modified Curie-Weiss law behavior<sup>35</sup> with  $T^*=2080$  K which is in-

dicative of strong hybridization in this compound. The  $5f$  electrons in  $\text{UGa}_3$  can be considered itinerant because of the large hybridization of  $5f$  orbitals with conduction electron orbitals. The photoinduced change in reflectivity, as shown in Fig. 5(a), can be fitted with a single exponential decay  $\Delta R/R(t) = A(T) \exp(-t/\tau)$ . The extracted relaxation amplitude  $A(T)$  and time  $\tau(T)$  are shown in Fig. 5(b) and Fig. 5(c), respectively. Upon entering the SDW phase,  $A(T)$  increases with decreasing temperature. However, instead of monotonically increasing as in  $\text{UAl}_3$  and  $\text{USn}_3$ ,  $A(T)$  now attains a maximum at  $\sim 40$  K and starts decreasing with decreasing temperature (see Fig. 5(b)). Concurrently,  $\tau$  exhibits a quasi-divergence at  $T_N$ , consistent with that observed in itinerant antiferromagnets  $\text{UNiGa}_5$  and  $\text{UPtGa}_5$ , where the opening up of the SDW gap causes a bottleneck in quasiparticle relaxation.<sup>14,15</sup> In contrast to  $\text{UNiGa}_5$  and  $\text{UPtGa}_5$ , however, where  $\tau$  increases with decreasing temperature at low temperatures,  $\tau$  in  $\text{UGa}_3$  shows a (1) shoulder (or change in curvature) at 40 K, and (2) decrease with decreasing temperature. An anomaly at a spin-reorientation temperature  $T_{sr}=40$  K has been reported in other measurements of  $\text{UGa}_3$ , whether in the presence of a magnetic field (nuclear magnetic resonance, neutron scattering, magnetic susceptibility),<sup>36–40</sup> or in the absence of a magnetic field (thermal conductivity and neutron scattering).<sup>36,37</sup> This anomaly has been associated with a reorientation of the ordered magnetic moments, which induces strong modifications of the uranium  $5f$  orbitals.<sup>39</sup> The fact that the transition is observed in the absence of magnetic field is an indication that the bump we see at 40 K in our pump probe measurement is not an artifact, but corresponds to the moment reorientation as has been reported in other measurements mentioned above.

We use the model proposed by Kabanov *et al.*<sup>41</sup> to analyze the temperature dependence of  $A$ . The temperature-dependence of the relaxation amplitude in the SDW state for an isotropic temperature-dependent gap  $\Delta_{SDW}(T)$  is given by (writing  $\Delta_{SDW}(T)$  as  $\Delta(T)$ )

$$A(T) = \frac{\epsilon_I / (\Delta(T) + k_B T/2)}{1 + \zeta \sqrt{\frac{2k_B T}{\pi \Delta(T)}} \exp[-\Delta(T)/k_B T]}, \quad (4)$$

where  $\epsilon_I$  the pump laser intensity per unit cell,  $\zeta$  is a constant, and  $\Delta(T)$  obeys a weak-coupling BCS temperature dependence. The above expression for  $A(T)$  describes a reduction in the photoexcited quasiparticle density with increase in temperature, due to the decrease in gap energy and corresponding enhanced phonon emission dur-

ing the initial relaxation. A good fit between the experimental  $A(T)$  and Eq. (4) can only be made from  $T_N$  down to  $\sim 40$  K, where  $T_N=55$  K is a fitting parameter. In the SDW state ( $T < T_N$ ), the temperature-dependence of  $\tau$  can be obtained from Eq. (3), but can be written in the alternative form (writing  $\Delta_{SDW}(T)$  as  $\Delta(T)$ )<sup>13</sup>

$$\tau^{-1}(T) = \Gamma \{ \delta A(T) + \eta \sqrt{\Delta(T)T} \exp[-\Delta(T)/T] \} \times [\Delta(T) + \alpha T \Delta(T)^4]. \quad (5)$$

The fit of  $\tau(T)$  to Eq. (5) is shown in Fig. 5(c). Once again, a good fit is obtained only from  $T_N$  to  $\sim 30$  K, close to  $T_{sr}$ . Below  $T_{sr}$ , the fit deviates from the experimental data, consistent with the existence of another transition at  $T_{sr}$ .

## V. CONCLUSION

We have performed time-resolved photoinduced change in reflectivity measurements on three isostructural uranium compounds —  $\text{UAl}_3$ ,  $\text{UGa}_3$  and  $\text{USn}_3$ . The values of  $T_{sf}$ , a measure of the degree of hybridization, in  $\text{UAl}_3$  and  $\text{USn}_3$ , are consistent with data from other measurements. Our fit of the slow component to the Rothwarf-Taylor model shows that the slow component can be described by assuming electron-hole pairs relaxing across the hybridization gap. We have thus shown the pump probe technique to be sensitive to c- $f$  hybridization. Our data on  $\text{UGa}_3$  is consistent with the formation of a SDW gap at  $T_N=60$  K, and a reorientation of magnetic moments at  $T_{sr}=40$  K.

## VI. ACKNOWLEDGEMENTS

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